Review

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Recent progress in photodetectors based on low-dimensional nanomaterials

<https://doi.org/10.1515/ntrev-2018-0084>

Received May 31, 2018; accepted August 13, 2018; previously published online September 12, 2018

Abstract: Photodetectors (PDs) have great potential in applications of imaging, telecommunication, and biological sensing. In this article, state-of-the-art achievements on typical low-dimensional nanostructured PDs and hybrid PDs are reviewed. In the 2D nanostructured PDs part, 2D transition metal dichalcogenides have a natural gap, which promise high sensitivity of photo-detection. Graphene and black phosphorus can also stand for 2D nanostructured PDs due to their broadband absorption and tunable direct bandgap, respectively. In the 1D nanostructured PDs part, owing to its high photoconductive characteristic, ZnO nanowire film is a promising material for ultraviolet PDs. Carbon nanotubes show potential in infrared (IR) detection due to its unique physical properties. In the 0D nanostructured PDs part, lead sulfide has a small bandgap and large Bohr exciton radius, which collectively give it a wide spectral tunability in the IR. In the hybrid PDs part, electrical and chemical doping is applied to combine different nanomaterials to realize PDs with high performance. In each part, the present situation and major challenges are overviewed. Then, the evolutions of the methods to overcome these challenges and the tremendous research breakthroughs are demonstrated. At last, future directions that could improve the performance of PDs are discussed.

Keywords: nanomaterials; photodetectors; typical low dimensional.

1 Introduction

During the past several decades, the booming photo-electronic industry has stretched throughout every facet of our lives. As one of the most critical optoelectronic devices, photodetectors (PDs), which can transform light into electrical signals precisely, have been stimulated into extensive research. Up to now, the accurate detection of light from the ultraviolet (UV; 10–400 nm) to the visible (400–760 nm) to the infrared (IR; 760–1 mm) to the terahertz (0.1–10 THz) region is critical for extensive and significant applications in military and civil use, such as night vision, imaging, sensing, and even information communication systems. Under the impetus of Moore's law, the feature size of highly integrated electronic devices has shrunk toward several nanometers. For example, Intel has achieved high volume production of 10 nm chips, and IBM Research Alliance produced the industry's 7 nm node test chips. Therefore, a major current focus in developing PDs is how to scale them down to the nanoscale for technological applications to realize a highly integrated photodetection system with high performance [1–4].

It has widely been shared that one way to improve the performance of PDs is to explore new low-dimensional nanomaterials [5]. The other way is to use hybridized structures. With one spatial dimension confined within the nanoscale, 2D materials, especially in terms of 2D layered materials and solution-processed nanofilms, exhibit many unique favorable properties for photodetection. Compared to the growth of conventional semiconductor films via universal epitaxial techniques, 2D materials are usually fabricated by simpler methods [6–8]. In addition, they have exceptional potential in electronic and optoelectronic applications [9]. For example, graphene exhibits extremely high charge-carrier mobility, optical transparency, and broadband absorption [10, 11]. In contrast to 2D nanostructured materials, having 1D limited within the nanoscale, 1D nanostructure materials possess an additional spatial dimension in total 2D confined within the nanoscale. As a result, they are ideal systems for exploring a large

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number of novel phenomena at the nanoscale as well as investigating the size and dimensionality dependence of properties for potential applications. In addition, its large surface-to-volume ratio and Debye length comparable to their sizes are also attractive. 0D nanostructured materials usually refer to materials whose excitons are confined in all three spatial dimensions (<100 nm). The nanoscale confinement of 0D materials makes it possible to tune its energy band by its size and shape. As a result of this 3D tunability of 0D materials, they usually exhibit several unique electronic and optical properties, especially for use of PDs [8, 9]. Hybrid method can help improve the performance of present low-dimensional PDs; for instance, to yield an ultrahigh responsivity of PDs, a special hybrid configuration consisting of 2D quantum dot (QD) materials was demonstrated [12]. The aim of this review paper is to present state-of-the-art PDs based on low-dimensional nanomaterials, including graphene, transition metal dichalcogenides (TMDCs), black phosphorus (BP), ZnO nanowire film, carbon nanotube (CNT), and lead sulfide (PbS). Moreover, the hybrid method incorporated in this field is demonstrated in detail as well. Here, the performance of PDs is measured by “5S” requirements: sensitivity, signal-to-noise ratio (SNR), spectral selectivity, speed, and stability. In the last part, the possible challenges and opportunities in the future development of these PDs are proposed.

2 2D nanostructured PDs

2.1 Graphene

Recently, graphene has emerged as an attractive material in PDs due to its high carrier mobility, broadband absorption, and mechanical flexibility. Due to graphene’s typical plasmonic nanostructures, photocurrent is no longer limited to photon energies greater than the bandgap of the semiconductor [9]. Moreover, it can be integrated into virtually any waveguide material, including Si, SiN, or AlN. Early data interpreted in the study have revealed an ultrafast photoresponse in graphene with an intrinsic bandwidth of 260 GHz, showing the promise for high-speed photodetection applications. Since then, several graphene-based PDs on Si waveguides have been realized [12].

Schuler et al. [13] presented an ultrafast graphene-based PD on an Si slot waveguide, where the Si strips serve as local back gate electrodes to create a p-n junction for efficient photodetection. A responsivity of 35 mA/W

at zero-bias conditions was achieved, and while under a moderate drain-source bias of 300 mV, the responsivity increased to 76 mA/W. This PD has shown a record high 3 dB bandwidth of 65 GHz. To further improve the responsivity in terms of V/W, the electrical gating could be extended closer to the contacts, for example, using thin Si slabs to both sides of the waveguide.

However, the realization of broadband PDs has been hindered by graphene’s low absorption in the visible and near-IR (NIR) regions. Kang et al. [14] demonstrated a stretchable PD based exclusively on crumpled graphene, which solved this barrier perfectly. As shown in Figure 1, the crumpled graphene, which has been intentionally deformed to have a continuously undulating 3D surface, induced an areal density increase to yield higher optical absorption per unit area, thereby improving photoresponsivity. As a result, this textured graphene PD exhibited a 370% enhancement in photoresponsivity compared to a flat graphene PD. More importantly, the stretching capability could reach up to 200% of its original length and no limit on detection wavelength. This surface engineering approach can be used to enhance photoabsorption and develop a highly responsive PD based on a single photoactive material of other emerging 2D materials such as TMDCs, including MoS_2 , WS_2 , and WSe_2 monolayers.

Fang et al. [15] demonstrated a broadband graphene PD exhibiting enhancement factors of 8 to 13 over the visible spectrum by reforming the source and drain contacts into a fractal metasurface and a ring encircling it. The metasurface was realized through a fractal tree to mimic the snowflake geometry, as shown in Figure 2. The application of this fractal metasurface was not bound to graphene PDs but could also be integrated with photovoltaic/photothermoelectric effect PDs made of other photodetection materials. The great flexibility of our fractal metasurface design enabled broadband enhancement at other portions of the electromagnetic spectrum and for various spot sizes. Additionally, the photoresponse enhancement was insensitive to the incident light polarization, a quintessential feature of a practical photodetecting/photoharvesting system. These attributes, combined with dynamic tunability through source-drain bias, make this fractal metasurface an advancement toward the incorporation of graphene into modern photodetecting and photoharvesting applications.

2.2 TMDCs

Great improvement in the responsivity has been realized for PDs based on graphene; however, there is an intractable

Figure 1: Mechanism of enhanced and strain-tunable photoresponsivity and photographs of textured graphene PD arrays. (A) Schematic illustrations of the physical mechanism enabling enhanced and strain-tunable photoresponsivity. Bottom schematic illustrations show close-up at the interface and illustrate the modulation of crumple areal density, height, and pitch in the illuminated region with an applied strain. (B) Photograph of a stretchable PD array (3*2 array). Bottom image shows an optical microscope image of the textured graphene PD. Reproduced with permission from Ref. [14].

Figure 2: Construction of the branching generated by recursive iterations. (A) Construction of the fractal design with "snowflake" geometry from levels 1-4. (B) Total structure comprising four-level (blue) and three-level (red) fractals used in the study. (C) Finite-difference time-domain simulated in-plane electric field (of the incident electromagnetic wave) distribution just underneath the Au fractal metasurface on a glass substrate under the excitation wavelength of 530 nm. The electric field is linearly polarized along the y-direction. Reproduced with permission from Ref. [15].

issue for graphene-based devices: the high dark current arising from its gapless nature, which has a strong impact on the sensitivity of photodetection [16, 17]. Alternatively, another kind of layered material, TMDCs with a natural gap, has captured much more attention in recent years. Up to now, TMDCs, including MoS_2 , WS_2 , WSe_2 , InSe , GaS ,

GaTe , GaSe , and In_2Se_3 [18], have been explored to develop plenty of new PDs with favorable photoelectronic properties. To illustrate this kind of PD vividly, MoS_2 -based PDs are taken as an example.

As a typical TMDC material, MoS_2 is composed of a vertical stack in which the weakly interacting layers are

held together by van der Waals forces. It is noteworthy that bulk-form MoS_2 is a semiconductor with an indirect bandgap of 1.2 eV, whereas single-layer MoS_2 is another kind of semiconductor with a direct bandgap of 1.8 eV [19]. This unique property of MoS_2 stems from the effect of quantum confinement on its electronic structure.

Ye et al. [20] demonstrated an advanced highly polarization-sensitive IR PD based on BP-on- WSe_2 van der Waals vertical heterostructure. This PD used a vertical photogate heterostructure of BP-on- WSe_2 in which BP serves as the photogate and WSe_2 as the conductive channel. Ultrahigh visible and IR photoresponsivity at room temperature can reach up to 1000 and 0.5 A/W, respectively, and ultrasensitive visible and IR specific detectivity is obtained up to 10^{14} and 10^{10} Jones, respectively, at room temperature. Moreover, the high sensitivity to IR polarization is about 40 mA/W with incident light polarized along the horizontal axis (defined as 0° polarization). It shows that the BP photogate structure of the BP-on- WSe_2 detector has higher sensitivity to polarized IR illumination photodetection and lower specific detectivity at room temperature. This detector provides a new strategy to integrate 2D layered materials to enable the corresponding PDs and may open up a new dimension for applications to IR imaging systems operating at room temperature.

Gomathi et al. [21] fabricated a novel paper-based broadband PD using hybrids of ZnS-MoS_2 . The PD was fabricated using a simple two-step hydrothermal method. The spectral selectivity of MoS_2 has been extended to UV wavelength region by combining MoS_2 with ZnS having high sensitivity toward UV light. The fabricated PD displays high sensitivity toward visible light when compared to UV and IR. The PD exhibits a responsivity of 4.5 $\mu\text{A/W}$ for IR, 9.4 $\mu\text{A/W}$ for UV light, and 17.85 $\mu\text{A/W}$ for visible light. In addition, ZnS-MoS_2 exhibited increased responsivity due to the reduced electron-hole recombination, which was a result of the straddling-type band alignment observed at the interface of ZnS-MoS_2 . This is the first report on paper-based broadband PD with ZnS-MoS_2 hybrids as active sensing materials and it provides a promising route for the development of large-scale paper-based broadband PDs using TMDC hybrids at low cost, having diverse applications in the field of wearable electronics, environmental monitoring, and surveillance.

2.3 BP

In addition to TMDC layered materials, BP is the latest developed layered material [22, 23]. After the groundbreaking research on BP in 2004 by Manchester

scientists, this layered material opened a new branch of science and provided a vast number of unlimited applications ranging from printable electronics to prototype devices [3].

In contact with other 2D nanomaterials, BP has intrinsic and tunable direct bandgap from 2.0 eV (monolayer) to 0.3 eV (bulk), which covers the optical spectrum range from visible to NIR. In addition, BP has also shown remarkable electrical properties, including a high hole mobility up to $1000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature, an on-off ratio up to 105, and good current saturation in field-effect devices [24]. BP-based field-effect transistors (FETs) and their performances are shown in Table 1. Furthermore, it has unique in-plane anisotropic physical properties unlike most of other 2D materials. The anisotropy of any physical property refers to the change of that property in different directions. It is not new in materials but is quite exciting in new 2D material BP due to its large magnitude in anisotropy compared to other 2D materials [3]. These characteristics make BP as the most promising candidate for broadband photodetection.

However, the current performance status of BP PDs in terms of broadband spectrum range, high photoresponsivity, and fast response time is still unsatisfactory. Furthermore, the impurities and the dielectric environment have an important impact on the electrical and optical properties of BP devices. In this section, we attempt to give a comprehensive overview and analysis of the published results of BP PD and show some new pathways to overcome these challenges.

Table 1: BP-based FETs and their performances.

Film thickness (nm)	Channel length (μm)	Gate insulator	Hole mobility ($\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)
10	1.6	SiO_2	984
5	1	SiO_2	286
5	1	SiO_2	205
18.7	3	SiO_2	170
5	0.69	SiO_2	155
1.6	0.45	SiO_2	35
8.5	0.3	HfO_2	400
1.9	2	SiO_2 (back)/ Al_2O_3 (top)	172
15	2.7	Polyimide (back)/ Al_2O_3 (top)	310
5			180
~5	–	Ionic liquid	510
8	–	h-BN	1350
10	–	h-BN	400

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Miao et al. [25] fabricated an NIR camera based on a single BP PD using the compressive sensing algorithm, which took advantage of the outstanding photoresponse of BP and overcame the difficulties of BP film growth and scalable PD array fabrication. The imaging was achieved by combining the PD with a digital micromirror device to encode and subsequently reconstruct the image based on compressive sensing algorithm. Stationary images of an NIR laser spot ($\lambda = 830$ nm) with up to 64×64 pixels were captured using this single-pixel BP camera with 2000 times of measurements, which was only half of the total number of pixels. The imaging platform demonstrated in this work circumvents the grand challenges of scalable BP material growth for PD array fabrication and shows the efficacy of using the outstanding performance of BP PD for future high-speed IR camera applications.

Ren et al. [2] synthesized few-layer BP nanosheets with large size by a facile liquid exfoliation method. Photoelectrochemical (PEC) tests demonstrated that the current density

of BP nanosheets can reach up to 265 nA cm^{-2} under light irradiation, whereas the dark current densities fluctuated near 1 nA cm^{-2} in 0.1 M KOH , as shown in Figure 3. In addition, the cycle stability measurement exhibited no detectable distinction after processing 50 and 100 cycles, whereas an excellent on-off behavior was still preserved even after 1 month. Furthermore, the PEC performance of BP nanosheet-based PD was evaluated in various KOH concentrations, which demonstrated that the as-prepared BP nanosheets might have a great potential application in self-powered PDs. The present work provides fundamental acknowledgment of the performance of a PEC-type BP nanosheet-based PD, offering extendable availabilities for 2D BP-based heterostructures to construct high-performance PEC devices.

Youngblood et al. [26] demonstrated a waveguide-integrated and gate-tunable BP PD integrated on an Si photonic waveguide operating in the NIR telecom band. In a significant advantage over graphene devices, these BP PDs could operate under bias with very low dark current and attained

Figure 3: Investigation of BP's optical property. (A) Schematic illustration of the proposed solution-phase exfoliation route for BP nanosheets. (B) Scanning electron microscopy (SEM) image. (C) EDS image. (D) Transmission electron microscopy (TEM) image. (E) High-resolution TEM (HRTEM) image. (F) Selected area electron diffraction (SAED) pattern of exfoliated BP. (G) UV-visible spectra of BP/NMP (inset: optical image of BP/NMP solution). (H) Raman spectra of bulk BP and exfoliated BP nanosheets on SiO₂/Si substrate. (I) Photoluminescence spectra of exfoliated BP nanosheets on SiO₂/Si substrate. Reproduced with permission from Ref. [26].

an intrinsic responsivity up to 135 and 657 mA/W in 11.5- and 100-nm-thick devices, respectively, at room temperature. The photocurrent was dominated by the photovoltaic effect with a high response bandwidth exceeding 3 GHz.

Deng et al. [27] demonstrated a gate-tunable p-n diode based on a p-type BP/n-type monolayer MoS₂ van der Waals p-n heterojunction. Upon illumination, these ultrathin p-n diodes showed a maximum photodetection responsivity of 418 mA/W at the wavelength of 633 nm and photovoltaic energy conversion with an external quantum efficiency of 0.3%. These p-n diodes show promise for broadband photodetection.

Miao et al. [28] demonstrated a truly vertically stacked and self-encapsulated van der Waals heterojunction diodes built from 2D semiconductors (MoS₂ and BP) and

compared its performance to conventional lateral 2D heterojunction devices (partially overlapped 2D heterostructures), as shown in Figure 4. Both vertical and lateral p-n heterostructure diodes exhibited a strong rectification ratio without gate voltage applied. Moreover, the current density under forward bias vertical diode delivered 70 times higher than a conventional lateral device design; it is attributed to the complete elimination of series resistance. This work demonstrates the potential of using the vertically stacked 2D semiconductors for future nanoelectronic and optoelectronic devices with optimal performance.

Li et al. [29] first fabricated a bipolar phototransistor based on WSe₂-BP-MoS₂ van der Waals heterostructure. Broadband photoresponsivities for visible (532 nm) and IR (1550 nm) light waves reach up to 6.32 and 1.12 A/W,

Figure 4: Fabrication procedure and characterization of vertically and laterally stacked BP/MoS₂ heterojunction p-n diodes. (A) Schematic diagrams illustrating the fabrication procedure of vertically stacked BP/MoS₂ p-n diodes. A 50 nm-thick Au film was used to fill the square trench followed by lift-off to form the bottom electrode (A-i). Few-layer p-type BP flake was then transferred onto the as-fabricated Au/p⁺-Si bottom electrode (A-ii). Next, the MoS₂ flake fully covers the BP (A-iii). Finally, the top electrode was patterned onto the MoS₂ flake by EBL and thermal evaporation of 50 nm-thick Au film (A-iv). The current can flow between the bottom Au/p⁺-Si electrode and the top Au electrode through the semiconducting BP/MoS₂ heterostructure (A-v). (B) Optical microscope images showing a representative vertical BP/MoS₂ p-n junction diode after each fabrication step including Au (B-i), BP (B-ii), MoS₂ (B-iii) and top Au electrode (B-iv). Scale bar, 10 μm. (C and D) Ambient stability study of (C) vertically and (D) laterally stacked BP/MoS₂ diodes. Scale bar, 10 μm (C-i and D-i) and 5 μm (C-ii, C-iii, D-ii, and D-iii). (E) Raman spectra of MoS₂, BP, and BP/MoS₂ heterostructure. Reproduced with permission from Ref. [29].

respectively, which are both improved by tens of times in comparison to similar photodiode devices composed of WSe_2 -BP. The phototransistor also exhibited ultrasensitive shot noise limit specific detectivities, which were 1.25×10^{11} Jones (1 Jones = $1 \text{ cm Hz}^{1/2} \text{ W}^{-1}$) for visible light at wavelength $\lambda = 532 \text{ nm}$ and 2.21×10^{10} Jones for the NIR light at wavelength $\lambda = 1550 \text{ nm}$ at room temperature. This generic strategy by combining 2D layered materials into heterostructures to enable multifunctional devices and components may open a new avenue for high-density monolithic integration of functional devices in a limited area.

Ding et al. [30] constructed a novel 2D van der Waals heterostructure made by InSe and BP monolayer with high hole mobility ($10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and investigated its structural and electronic properties using first-principles calculations. The InSe/BP heterostructure could be formed easily due to its negative binding energy. Its heterostructure exhibited an intrinsic type II band alignment with a direct bandgap of 1.39 eV, in which electrons were located in the InSe layer and hole localized in the BP layer. The band offsets of InSe and BP are 0.78 eV for CBM and 0.86 eV for VBM, respectively, which greatly enhance the performance of InSe-based photovoltaic devices and PDs. The electron mobility of the InSe/BP heterostructure is increased to $3 \times 10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and hole mobility is increased from $40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in InSe monolayer to $10^4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is three orders of magnitude larger than that in InSe monolayer. Thus, the InSe/BP heterostructure is considered to have a huge potential application in PDs.

Ye et al. [31] demonstrated a PD with visible to NIR detection range based on the heterojunction fabricated by van der Waals assembly between few-layers BP and few-layers MoS_2 . The heterojunction with electrical characteristics, which could be electrically tuned by a gate voltage, achieves a wide range of current-rectifying behavior with a forward-to-reverse bias current ratio exceeding 10^3 . The photoresponsivity of the PD was about 22.3 A/W measured at $\lambda = 532 \text{ nm}$ and 153.4 mA/W at $\lambda = 1.55 \mu\text{m}$ with a microsecond response speed (15 μs). In addition, its specific detectivity was calculated to have the maximum values of 3.1×10^{11} Jones at $\lambda = 532 \text{ nm}$ and 2.13×10^9 Jones at $\lambda = 1550 \text{ nm}$ at room temperature. This generic strategy to integrate 2D layered materials in the vertical direction to enable functional devices and circuits may open up a new dimension for high-density integration of functional devices in the limited circuit area.

In addition, BP can also be configured as an excellent UV PD. Wu et al. [32] investigated a UV PD based on BP with a specific detectivity of 3×10^{13} Jones for the first time. The UV photoresponsivity could be significantly enhanced to $9 \times 10^4 \text{ A/W}$ by applying a source-drain bias

of 3 V, which was the highest ever measured in any 2D materials and 10^7 times higher than previously reported value for BP. Such a colossal UV photoresponsivity was attributed to the resonant-interband transition between two specially nested valence and conduction bands. These nested bands provide an unusual high density of states for highly efficient UV absorption due to their singularity nature. This work shows that BP proves to be a strong viable candidate for future optoelectronic applications especially as state-of-the-art UV detectors.

3 1D nanostructured PDs

3.1 ZnO nanowire film

ZnO is a group II to VI wide-bandgap semiconducting metal oxide with high electron mobility and exciton binding energy, which makes it an interesting material for a variety of applications [33, 34]. Owing to their wide bandgap (3.4 eV) [35], high exciton binding energy (60 meV), and high photoconductive characteristic, ZnO nanostructures are frequently used to construct UV PDs [36–39]. ZnO films with high specific area and physical flexibilities are easy and cheap to prepare and process with low costs, and the photoconductivity of ZnO film PDs changes notably upon on-off state switching, demonstrating their potential in practical applications [37, 40]. In particular, forming heterojunctions between ZnO and other materials [ZnS, poly(vinyl alcohol), poly(acrylonitrile), and poly(vinylcarbazole)] has significant effects on improving the photoelectric performance of ZnO film-based PDs [41–43]. For instance, ZnO- Ga_2O_3 core-shell microwire PD is highly suitable for practical applications of solar-blind photodetection [44].

A novel CdMoO_4 -ZnO composite film has been prepared by spin-coating CdMoO_4 microplates on ZnO film and was constructed as a heterojunction PD. With the aid of CdMoO_4 microplates, the structure of the PD changed from MSM type to a heterojunction one, which largely benefited the photoelectric performance. Ouyang et al. [37] deposited Au nanoparticles (NPs) to modify the CdMoO_4 -ZnO composite film, and compared to the CMO-ZnO PD, this PD achieves a higher photocurrent of 289.1 nA and a higher responsivity of 321.1 mA/W. At the same time, its decay time decreases to 9.2 s. This further improvement of photoelectric properties was due to the hot electron injection effect of AuNPs and the Schottky barriers formed between the AuNPs and CMO-ZnO composite film block the carrier transportation. However, an excess loading

of CdMoO_4 microplates might shelter the ZnO film underneath and block the light absorption of the composite film, which would lead to the decrease of photocurrents. Thus, in practical applications, it should be avoided. The study on Au-CMO-ZnO composite film PD might be potentially useful for facile construction of ZnO-based PD with novel structure and high photoelectric performance.

Teng et al. [42] incorporated a BiOCl nanostructure into the ZnO film and constructed them as film PDs. With forming barriers to block the straight transmission of electrons between electrodes, the rise/decay time of BiOCl/ZnO hybrid film PD was 25.83/11.25 s, which was much shorter than that of pure ZnO film PD (69.34/>120 s). Due to the injection of photogenerated electrons into the ZnO film under UV light illumination, the responsivity of the BiOCl/ZnO hybrid film PD in the UV region could reach 182.87 mA/W, which was about 6.87 times that of pure ZnO film PD. This is different from those methods that enhanced the performance of PDs by adjusting the depletion layer on the surface of ZnO NPs. The BiOCl/ZnO hybrid structure film PD provides a new and simple route for the enhancement of optoelectronic performance based on the ZnO nanostructures.

Duan et al. [45] fabricated a ZnO:Al/Si heterojunction PD by a simple and low-cost CBD method. ZnO:Al/Si PD showed a sharp cutoff responsivity at 400 nm, a UV-to-visible rejection ratio ($R_{386\text{ nm}}/R_{500\text{ nm}}$) of 50, and fast rise and decay times of the photoresponse, which indicated that the hexagonal nanocrystal (NC) ZnO:Al film was the most promising materials for the fabrication of high-performance UV PDs. Self-powered UV PDs, which have vast applications in the military and for civilian purposes, have become particularly attractive in recent years due to their advantages of high sensitivity, ultrasmall size, and low power consumption. This should generate extensive interest in this field and encourage more researchers to engage in and tackle the scientific challenges [46–48]. In 2017, Yu et al. [49] fabricated three novel heterojunctions by depositing films through one-step *in situ* polymerization in acid solution, which provides a simple and efficient approach to prepare high-performance self-power UV PDs. Hu et al. [39] developed a high-performance UV PD based on an Se/ZnO p-n heterojunction. This device was theoretically equal to a parallel-connection circuit for its special structure, and due to its parallel connection of ZnO and the Se/ZnO hybrid structure, this UV PD realized a binary response of positive and negative output currents using an on-off light source at a small reverse bias. Additionally, the speed of the ZnO UV PD is greatly improved with a rise time of 0.69 ms and a decay time of 13.5 ms, which are much quicker than those of pure ZnO film PDs.

All the results demonstrate that the high-performance UV PD based on an Se/ZnO p-n self-powered heterojunction provides a promising way to fabricate ZnO-based p-n junction UV PDs with high sensitivity, high SNR, high spectral selectivity, high speed, and high stability and thus may lay a solid ground for the future applications of this kind of high-performance multifunctional PD.

Purusothaman et al. [50] synthesized highly crystallized ZnO microarchitectures (ZnO microwire, ZnO coral-like microstrip, and ZnO fibril-like clustered microwire) through a simplified vapor transport technique (carrier gas/catalyst-free). As-fabricated PD is shown in Figure 5. Among them, ZnO fibril-like clustered microwire exhibited higher IP_{h290} μA at 17 mW cm^{-2} and comparable responsivity ($R_{405\text{ nm}}$) of 716 mA/W. In contract to reported microarchitectures, ZnO fibril-like clustered microwire exhibited superior photosensing behavior with a detectivity range of 6.72×10^{10} Jones and broadened spectral activity extending from UV to the visible region (365, 405, and 535 nm). The results reported in this paper provide a facile route to develop flexible self-powered optical communication systems and sensors.

Chen et al. [51] developed a strain modulated solar-blinded avalanche PD based on ZnO- Ga_2O_3 core-shell heterojunction microwire. This PD is highly sensitive to deep UV light centered at 261 nm. Due to its ultrahigh sensitivity and spectral selectivity, this PD could respond to rare weak deep UV light with almost no response to visible light. Moreover, the detection could be enhanced by applying certain static strains on the device through the piezo-phototronic effect. The current response could be enhanced about three times under -0.042% strain. The strain-induced piezopotential modulates carrier transport mainly in the ZnO core. The photoresponse of this PD was enhanced by this effect and thus made it a more perspective PD in human-machine interaction areas, such as health monitoring, smart systems, and security communication.

3.2 CNTs

1D semiconductor nanostructures have stimulated much attention in high-performance PDs, owing to the pronounced surface effects originating from the large surface-to-volume ratio [52, 53]. Since the discovery of multiwalled CNT by Iijima and Ichihashi and the related single-walled CNTs in 1993 [54], CNTs exhibited some strong advantages for applications in IR detectors in comparison to other 1D candidate materials, e.g. the naturally nanoscale size, simplified growth and device fabrication, chirality-dependent

Figure 5: Structure and photospectrum of this as-fabricated ZnO. (A) Raman spectrum of ZnO microarchitectures. (B) Schematic representation of as-fabricated PD. (C) Compressive strain-induced behavior of PD. (D) Schematic view of ZnO device architectures. (E) Optical images of as-fabricated PD devices using ZnO MW, ZnO CMS, and ZnO F-MW. Reproduced with permission from Ref. [50].

direct band structures that cover important communication wavebands, and room-temperature operation due to their moderate bandgap energies ($\gg 26$ meV) [55]. The very small size and good extendibility also provide great prospects for their flexible integrations into other photonic or hybrid optoelectronic systems [56, 57].

However, for CNT-based near- or shortwave-IR optoelectronics, the existing developments for enhanced absorption or signal detection are insufficient [58]. Besides, for signal sensing or information imaging applications, which involve the detection of a specific target signal among a broad background of signals or noise, the photoresponse of CNT aggregations usually covers a wide NIR scope with various characteristic peaks induced by chirality-dependent Van Hove singularity transitions [59], leading to random responses without any targets. That is to say, a specific response to a target signal requires a higher suppression ratio [60]. To further increase the signal outputs, the hybrid systems that enhance light absorption can also be considered. Compared to the fast progress in the fields of 2D materials [61], some concepts for enriching CNT PDs should be further investigated and developed. Here, we present a figure of merit and some enhanced light absorption PDs in this section.

In 2016, Liang et al. [55] demonstrated a high photoelectric conversion efficiency and signal recognition CNT PDs by integrating with a designed F-P cavity. A ~ 6 -fold enhanced optical absorption could be achieved because of the confined effect of the designed optical mode, and the detectors exhibited a higher suppression ratio until a power density of 0.07 W cm^{-2} and photocurrent of 5 pA, and the spectral full-width at half-maximum is ~ 33 nm at a signal wavelength of 1200 nm. More importantly, a concept of the “resonance and off-resonance” cavity was developed at signal detection at zero bias, which opened the possibility of chirality-specific CNT film-based signal recognition systems for information photonics on a chip and broadening the potential for the application of other nanomaterials in information photonics regardless of their doping polarity. The concept offered a promising and practical pathway for the development of stable, large-scale, multiwavelength signal capture, recognition, and sensing systems.

Pyo et al. [62] demonstrated a heterogeneous all-carbon PD composed of graphene electrodes and porphyrin-interfaced single-walled CNTs channel, exhibiting high photoresponse, flexibility, and full transparency across the device. The porphyrin molecules generate

and transfer photoexcited holes to the single-walled CNTs even under weak white light, resulting in significant improvement of photoresponsivity from negligible to $1.6 \times 10^{-2} \text{ A W}^{-1}$. Simultaneously, the PD exhibited high flexibility, allowing stable light detection under $\approx 50\%$ strain (i.e. a bending radius of $\approx 350 \text{ }\mu\text{m}$) and retaining a sufficient transparency of $\approx 80\%$ at 550 nm . Notably, minimal loss in photoresponse even after heavy crumpling (a strain of $\approx 50\%$) and ability to detect sunlight gave the device great potential as an epidermal sunlight sensor. This PD could be used for low-cost, large-scale, and high-performance optoelectronic applications that require mechanical flexibility and optical invisibility. The hybridization of low-dimensional nanomaterials envisions a promising strategy for achieving flexible and transparent electronics as well as enabling further exploration into the unprecedented functionalities of nanomaterials.

However, the absorption restriction from single layer limits the effective utilization of incident light. In 2017, Huang et al. [63] demonstrated a plasmonic electrode structure in CNT thin-film PD based on randomly deposited high-purity semiconducting CNT, which could collect photoinduced carriers effectively and enhance light absorption at the same time. The plasmonic enhancement resonate around 1650 nm wavelength well corresponding to our simulation, which was a short-wave IR for plasmonic enhancement application. Furthermore, the best performance improvement of CNT detector with plasmonic structure could be enhanced as 13.7 times for photocurrent mode and 5.62 times for photovoltage mode than those devices without structure at 1650 nm resonant wavelength. At last, the plasmonic structures were applied on tandem PDs with nine virtual contacts, and both the photocurrent and the photovoltage were increased. By further improving the structure and the quality of CNT film, such as dense semiconducting CNTs array with thicker layer, the plasmonic detectors will show more powerful use in modern room temperature IR detection and even in optical integration circuit with compact construction.

4 0D PbS QDs

PbS has been among the most intensively investigated as a candidate material for solution-processed optoelectronics [64, 65]. This is primarily due to its small bandgap and large Bohr exciton radius, which collectively give it wide spectral tunability in the IR [66]. Recent efforts employing micron-sized ultrathin 2D PbS nanoplates as the active

material in PD devices showed efficient detection in the IR [67]. However, these nanoplates were fabricated via a chemical vapor deposition process that can be expensive and challenging to scale up in terms of production volume [68]. In 2010, Schliehe et al. [69] introduced the wet-chemical synthesis of 2D PbS nanosheets based on the 2D oriented attachment of sphere-like PbS particles into ultrathin planar structures. Their approach was subsequently adopted by other researchers who explored different synthesis parameters that allowed for shape and thickness control. Aside from oriented attachment, nucleation and growth within a lamellar octylamine bilayer template has also been shown to produce pseudohexagonal PbS quantum plates. Despite theoretical predictions of improved carrier mobility and stronger photon absorption [70], a thin film of nanosheets produced via the method of Schliehe et al. [68] yielded relatively low responsivity when used within a PD configuration. Additionally, we found that these nanosheets are prone to severe aggregation upon processing and cannot easily be separated into isolated sheets even after prolonged sonication. These limitations prompted us to develop an alternative approach to the synthesis of 2D PbS NCs. Here, a new solution-processed PbS QD was fabricated and applied for PDs.

Wu et al. [67] reported the synthesis of colloidal 2D PbS nanoplates with residual Cu_2S domains via a partial cation-exchange process involving Pb^{2+} and presynthesized hexagonal Cu_2S nanoplates with an average thickness of 3 nm and edge lengths of 150 nm . Due to the low resistivity of $8.04 \text{ }\Omega \text{ cm}$ of the PbS nanoplate film, which was comparable to commercial doped semiconductors, the responsivity and specific detectivity at 808 nm was 1739 A/W and $2.55 \times 10^{11} \text{ Jones}$, respectively. This work uses partial cation exchange and plate-like morphology in colloidal PbS NC film to improve its in-plane conductivity while retaining good photoresponse, and it needs to be investigated in much more detail how better photodetection efficiencies may be expected with higher NC packing densities and a more judicious choice of ligands, making these heterostructured PbS NPL films possible candidates for low-cost high-performance solution-processed NIR PDs.

Shen et al. [71] demonstrated a highly sensitive, visible-blind, and narrow-band NIR PD with a full-width at half-maximum of 50 nm and a visible rejection ratio of 100. This device had a thick nanocomposite absorbing layers made of PbS QDs, which effectively enhanced both the responsivity and the visible rejection ratio of external quantum efficiency by 10-fold compared to those without PbS QDs. Encouragingly, the inclusion of PbS QDs does not increase the device noise. We directly measured a noise equivalent power of 6.1 pW cm^{-2} at 890 nm

and a large linear dynamic range of more than 11 orders of magnitude. The highly sensitive visible-blind NIR narrow-band PDs may find applications in biomedical engineering.

Lee et al. [72] fabricated a solution-processed inorganic UV-visible-SWIR PD with light sensitivity from 350 to 2000 nm using highly monodispersed large PbS NCs with extremely low size dispersions (5%). These highly monodispersed large PbS NCs were synthesized by multiple injection of the sulfur precursor solution to grow the NCs without initiating new nucleation during growth, which enabled PbS NC synthesis with excellent particle size control and resulted in well-defined absorption peaks at long wavelengths. This UV-visible-SWIR multispectral PD showed detectivity values of more than 1×10^{11} Jones from 350 to 2000 nm and a maximum detectivity value of 1.2×10^{12} Jones at the peak wavelength of 1800 nm. This work realized the synthesis of large PbS NCs with low cost and practical applications.

Qiao et al. [73] demonstrated spectra-selective PbS QD IR PDs using the semiconductor's "filter" absorption characteristics and QD quantum size effect. Different from the wide detection spectra of traditional PDs, this PbS QD PDs can detect the spectra-selective light source. Spectra-selective PDs of perovskite/QDs and QDs/QDs were respectively implemented by integrating two functional layers. The top layer (facing the light) was used to filter the nontarget spectra and the bottom layer was used for detecting. When tuning the QD size, the response spectrum wavelength and range of spectra-selective PDs could be conveniently tailored. The obtained selectivity factor and normalized detectivity ratio from target and nontarget illumination can reach at least 10. These favorable properties promised spectra-selective PDs to be widely applied in identifying an unknown light source and future artificial recognition, artificial intelligence, communication, and surveillance system.

Mishra et al. [74] fabricated NIR PDs with a very simple architecture comprising a PbSe/PbS tetrapod layer between two Au electrodes on a glass substrate. PbSe/PbS tetrapod was synthesized by the cation exchange method. Due to its branched morphology, when illuminated by an NIR laser, these devices achieved a responsivity of 11.9 A/W without the use of ligand-exchange processes, thermal annealing, or hybrid device architecture. This heterostructured PbSe seeded PbS nanotetrapods was the most promising material of NC-based NIR PD. More importantly, the use of cationic exchange processes to derive anisotropic semiconductor nanoheterostructures can result in a fortuitous combination of properties that may be highly desirable for solution-processed optoelectronic applications.

Lee et al. [75] demonstrated high gain and low dark current solution-processed colloidal PbS QDs IR PIN PDs with IR sensitivity up to 1500 nm. At a peak wavelength of 1200 nm, the PD had a gain of 187, a detectivity of 7.0×10^{13} Jones, and a response time of 3.0 ± 0.2 ms. Due to its higher detectivity than commercial InGaAs PD, the sufficient response time for imaging applications, and the potentially low-cost fabrication process, this kind of IR PDs was promising for low-cost IR sensing applications [52].

Recently, a new kind of semiconductor, halide perovskite, has attracted worldwide attention. Intrinsically high mobility and large absorption coefficient endow halide perovskites with great promise for high-performance PDs [76–78]. Liu et al. [79] demonstrated solution-processed ultrasensitive broadband PDs using $\text{CH}_3\text{NH}_3\text{PbI}_3$ and PbS QDs as the light harvesters. When passivating the structural defects on the surface of PbS QDs with diminutive molecular-scaled $\text{CH}_3\text{NH}_3\text{PbI}_3$, the dark current of the PbS QDs/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ -based PDs were tremendously suppressed, which effectively brought down the recombination losses and consequently raised the photoresponse of the PDs. As a result, PDs exhibited a broad spectral response from 375 to 1100 nm, with high responsivity of more than 300 and 130 mA/W and high detectivity larger than 10^{13} and 5×10^{12} Jones in the visible and NIR regions. The results offered a facile and promising route for advancing the performance of broadband PDs.

However, when integrating PDs with metal NPs, the improvement of the overall device performance is severely hampered. In 2017, Ren et al. [80] fabricated a bilayer QD-only PD. Due to the QD junction controlled photo-carrier dynamics, upon light on, the junction greatly facilitates carrier separation at the interface and, upon light off, the junction promotes the recombination of accumulated carriers in the two films via the interface. The bilayer device is superior over the conventional single-layer devices with faster light response, broader linear dynamic range, larger detectivity, and higher SNR along with comparable responsivity. In addition, the bilayer device was successfully fabricated on flexible PI substrate with comparable performance to the glass-based one, signaling great potential of the QD-based bilayer PDs in future flexible optoelectronics.

Wen et al. [81] developed a high-performance IR PD by integrating nonlayered PbS and layered MoS_2 nanostructures via van der Waals epitaxy. The morphology is shown in Figure 6. Density functional theory calculations indicated that PbS nanoplates were in contact with MoS_2 edges through strong chemical hybridization, which was expected to offer a fast transmission path for carriers that enhanced the response speed. The phototransistor

Figure 6: Morphology of nonlayered PbS nanoplates-MoS₂ heterostructures. (A) Schematic illustration of PbS nanoplates-MoS₂ heterostructures. (B) Optical microscopy image with selectively growing PbS nanoplates along the edge of MoS₂ triangle. Inset is an amplified SEM image of edge contact heterostructures. (C) Atomic force microscopy image of edge of MoS₂ triangle. (D) HRTEM image with crystal fringes of (200) planes. Inset shows the SAED cubic pattern. (E) and (F) exhibit the statistics of the lateral dimension distribution and thickness distribution of the PbS nanoplates on MoS₂. The histogram statistics of PbS nanoplates: the lateral dimension and thickness distribution ranging from 150 to 750 and 20 to 80 nm, respectively. Reproduced with permission from Ref. [81].

exhibited a fast response as well as high photoresponsivity (4.5×10^4 A/W) and $I_{\text{light}}/I_{\text{dark}}$ (1.3×10^2) in the NIR spectral region at room temperature. In particular, the detectivity was as high as 3×10^{13} Jones, which was even better than that of commercial low-layered PDs. Furthermore, by controlling the growth and microfabrication patterning, periodic device arrays of PbS-MoS₂ that were capable of IR detection were achieved on Si/SiO₂ substrates. This work provides a possible method for the integration of PD arrays on Si-based electronic devices and lays a solid foundation for the practical applications of PbS-based devices in the future.

Liu et al. [82] reported solution-processed PDs fabricated by CH₃NH₃PbI₃ incorporated with PbS QDs through the trap-assisted charge-injection effect. The structure of this device is shown in Figure 7. PbS QDs, which possessed large amount of trap states, were introduced into CH₃NH₃PbI₃ thin film for establishing ohmic contact at the CH₃NH₃PbI₃/Al interface, realizing the elimination of the

charge injection barrier from the cathode electrode. As a result, an external quantum efficiency of 4500, a photoresponsivity of 15,000 mA/W and a detectivity of more than 6×10^{13} Jones at a small bias of 2 V, and a photoreponse time of 11.5 μ s were observed from the solution-processed PDs fabricated by the CH₃NH₃PbI₃:PbS QD nanocomposite thin film.

Owing to their ease of fabrication, low cost, and high flexibility, organic materials have attracted great interest in PD applications. However, suffering from large dark current, small photocurrent, low on-off ratio, and low sensitivity, performances of bare organic-based PDs are not satisfactory. Integrating organic materials with other novel semiconductor materials offers an opportunity to overcome these drawbacks. Wei et al. [83] demonstrated a lateral hybrid organic/PbS QD bilayer PD. Under light illumination, photogenerated electrons and holes are spatially separated via the junction and are transported in the PbS-TbAl QD layer and the organic layer, respectively.

Figure 7: Device structure and proposed working mechanisms of perovskite hybrid PDs (pero-H PDs). (A) Device structure of pero-H PDs and energy level alignment of the materials used for the fabrication of pero-H PDs (B) in dark and (C) under illumination. Reproduced with permission from Ref. [82].

This largely reduced carrier recombination and leads to enhanced photocurrent, and the dark current was greatly suppressed due to the formed organic blend/PbS heterojunction. The bilayer PD significantly outperforms the organic PD and the PbS PD and exhibits high photo-response of 6.32 A/W, large SNR of 1690, and high detectivity of 1.12×10^{13} Jones. The bilayer PD fabricated on flexible PI substrate shows comparable performance to the device made on glass substrate, signaling its great potential in future flexible optoelectronics.

5 Doping

Most reported PDs are built from inorganic materials such as ZnO, CdS, ZrS₂, and InAs [84–89]. To improve the performance of PDs, doping techniques are now turning into a sizzling area. Take graphene, for instance; the doping in graphene can be classified into electrical and chemical doping [90]. The doping is referred to as the chemical one when the changes in the graphene lattice structures result via chemical routes, such as substitutional doping with heteroatoms or molecules [91–93]. The goals achieved from such modifications strongly

depend on the type of dopants and concentrations and their location within the graphene systems, the chosen sites of graphene for replacement with the foreign impurity atoms, are of critical importance where Fermi level depends on the concentration of doped holes and electrons [83, 94].

Integrating plasmonic nanostructures with graphene demonstrated the possibility of effectively enhancing optical absorption by a strong field enhancement. In 2011, PDs with the AuNP array on graphene, which exhibited 1500% higher photoresponsivity compared to that without the AuNP array, were reported. The photoresponsivity was plasmonically enhanced by the excited surface plasmons confined in the plasmonic nanostructure. Although the integration enabled high photoresponsivity, most graphene-based PDs with the hybrid approaches still have limited stretchability. Recently, Kim et al. [95] reported a stretchable PD based on the hybrid structure of graphene and graphene QDs with high photoresponsivity. However, their PD has a limited stretchability only up to 25. Furthermore, mechanical robustness was demonstrated only up to 30 cycles of mechanical stretching.

Ni et al. [96] demonstrate the highly sensitive MIR photodetection of QDs/graphene hybrid phototransistors

Figure 8: Schematic and band energy of ZnO nanofiber. (A) Schematic of ZnO nanofiber and perovskite hybrid PDs. (B) Band energy alignment between ZnO and perovskite. Reproduced with permission from Ref. [88].

using plasmonic Si QDs doped with boron. The localized surface plasmon resonance enabled the MIR photodetection of QD/graphene hybrid phototransistors of boron-doped Si QDs, and the photogating effect following the band-to-band and sub-bandgap optical absorption of boron-doped Si QDs caused QD/graphene hybrid phototransistors to effectively operate in the UV-to-NIR region. Therefore, ultrabroadband UV-to-MIR photodetection of QD/graphene hybrid phototransistors with high sensitivity was realized. The current work has important implications for the development of high-performance optoelectronic devices based on the hybrid structures of 0D plasmonic QDs and 2D materials such as graphene.

Cao et al. [88] fabricated a hybrid PD consisting of electrospun ZnO nanofibers and perovskites for the first time. As shown in Figure 8, ZnO nanofibers were synthesized on indium-tin oxide glass with etched gap by the electrospinning method and then the perovskite layer was deposited onto the ZnO by a solution processing technique. Compared to pristine ZnO or perovskite, this hybrid PD showed increased on-off ratio, faster response speed, and higher responsivity and detectivity. The significantly improved performance was attributed to the fact that photogenerated charges were separated efficiently, owing to the matched energy band levels between ZnO and perovskite. The results provide a new and promising route to integrate inorganic functional materials with perovskite for high-performance and low-cost PDs.

Fu et al. [97] prepared a novel AgD/AgCl hybrid through a simple electrodeposition method using AgNO_3 and CTAC and precursors. The prepared AgD/AgCl hybrid consisted of Ag dendrites and AgCl polyhedral. Energy-dispersive X-ray spectroscopy (EDS) analysis and X-ray diffraction (XRD) characterizations confirmed that the

hybrid synthesized through electrodeposition had high purity. The prepared AgD/AgCl hybrid with a specific morphology exhibited an effective photosensitive property with a fast response time and good photocurrent reproducibility.

Gogurla et al. [98] demonstrated ZnO nanorod hybrid PDs on AuNP-embedded silk protein for flexible optoelectronics. Hybrid samples exhibited optical absorption at the band edge of ZnO as well as plasmonic energy due to AuNPs, making them attractive for selective UV and visible wavelength detection. The device prepared on Au-silk protein showed a much lower dark current and a higher photo to dark current ratio of 10^5 compared to the control sample without AuNPs. The hybrid device also exhibited a higher specific detectivity due to higher responsivity arising from the photogenerated hole trapping by AuNPs. The proposed novel hybrid ZnO nanorods on silk protein can provide a pathway for new opportunities toward low-cost, biocompatible, biodegradable, flexible, and lightweight PDs.

Ouyang et al. [37] reported a novel hybrid structure of BiOCl/ZnO film and TiO_2 /ZnO film by a facile spin-coating method and constructed it to PDs. As shown in Figure 9, due to the formation of p-n junction between the BiOCl and ZnO, compared to the obtained TiO_2 /ZnO hybrid film PD, the BiOCl/ZnO film PD exhibits excellent optoelectronic properties, including higher photocurrent/responsivity and shorter response time, outperforming the pristine ZnO film PD counterpart. However, excess BiOCl clusters in the hybrid film would significantly block the transport path of electrons between electrodes, resulting in the decrease of photocurrent. Thus, we should put emphasis on how to develop the construction of BiOCl/ZnO hybrid film to increase photocurrent.

Figure 9: X-ray photoelectron spectroscopy of different films: (A) XRD patterns of the ZnO film (black line), TiO_2 NPs (red line), and BiOCl nanosheets (blue line). (B) Survey scan. (C) $\text{Bi} 4f$. (D) $\text{Ti} 2p$ spectrum. Reproduced with permission from Ref. [42].

Lee et al. [99] demonstrated the fabrication of a novel hybrid PD consisting of graphene and $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite layers. The device exhibited a broad spectral photoresponsivity between 800 and 400 nm, and the photoresponsivity and effective quantum efficiency were 180 A/W and 5×10^4 at a relatively high illumination power of 1 μW , respectively. All of these high performances were attributed to the efficient charge transfer from the graphene to the perovskite. However, when this device showed fast rise time in the photoresponse, the relatively slow fall time hindered the performance of the PD. This problem might be solved by introducing an additional layer (on top of the device) having a lower valence band than $\text{CH}_3\text{NH}_3\text{PbI}_3$. The hybrid PD described here is expected to contribute to the development of imaging sensors suitable for low-light photography, UV detectors with a high responsivity, and smart skin sensors combined with other functionalities.

6 Conclusion

This review summarizes the latest research on PDs based on low-dimensional nanomaterials. Nanostructured materials with a number of fascinating and excellent photoelectric properties have promised themselves as versatile building blocks in terms of interconnects and functional units for novel PDs. Therefore, they are expected to play an important role in improving the performance and extending the function of traditional PDs.

However, with regard to the overall perspective, the study of nanoscale PDs is still at its preliminary stage. To make full use of nanostructured materials' unique photoelectric properties for practical applications, studies of many issues need to be urgently intensified.

In view of the basic scientific research and potential technological applications, a roadmap for further directions of nanoscale PDs is described as follows. First, the

ability to control nanostructured materials precisely is essential for high-performance nanodevices. Second, studies focusing on improving the photocurrent of hybrid PDs is needed. Additionally, to further increase the quantum efficiency of the photodetection, a new design of nanostructures with high efficient absorption of the incident light and hot electron generation is preferred. Last but not least, the use of new materials such as low-dimensional materials will also extend the spectral range of light absorption and hot-electron generation, allowing efficiently converting the light to current signals.

Although many riddles need to be solved, behind this burgeoning field, it is no doubt that the combination of well-studied materials and established device fabrication technologies will lead to more and more exciting discoveries in the further development of PDs. In view of the complexity of researching and developing nanoscale PDs, research tools from different disciplines, including physics, chemistry, materials science, and microelectronics, need to be pulled together to solve these problems. Of course, challenges mean opportunities as well, the entirely new era of photodetection is beckoning us on, and we do hope this review will inspire an increasing number of researchers and engineers to join this game.

Acknowledgments: This work was supported by the National Nature Science Foundation of China (Grant No. 61604019) and the National Natural Science Foundation of Liaoning (Grant No. 20170540748).

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